High Temperature Removal of H_2S from Coal Gasification Process Streams Using an Electrochemical Membrane System

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An electrochemical cell to be used for desulfurization of coal gasification process streams is being tested at lab scale. Syn-gas is passed by the cathode of the cell, run at typical gasifier temperatures, where the H_2S is preferentially reduced to hydrogen gas, which enriches the fuel gas, and sulfide ions, which enter the electrolyte-filled membrane. These sulfide ions are oxidized to elemental sulfur vapor at the anode electrode, collected downstream, providing continuous performance. A gas mixture reflective of coal gasification process streams is fed into the electrochemical cell, which is kept at temperature by a custom-made furnace. At 600• C, after hydration and shift reaction, the fuel gas composition entering the cell is 34% CO, 22% CO_2, 35% H_2, 8.5% H_2O, and 450 ppm H_2S. Gas chromatography is used to monitor H_2S concentrations into and out of the cell. Infrared spectroscopy is used to monitor CO_2.

The key to optimum performance is the cathode material, which is exposed to the sour gas. Previous work under UCR sponsorship has shown that nickel and cobalt electrodes are operational but have some limitations in H_2S concentration and temperature. We have recently tested a new ceramic material that appears stable and conductive over large ranges in concentration as well as temperature. Full-cell work has begun utilizing this material, as well as comparison with known materials.

Results will be presented for the performance of three different cathode materials: Co_9S_8, $LiY_{(0.9)}Ca_{(0.1)}FeO_3$, and NiS. Operation temperatures were varied from 580 to 650• C while system pressure remained constant at 1 atm. The other electrochemical cell components consist of a lithiated nickel anode, an inert ceramic membrane (yttria-stabilized zirconia), steel housing (passivated by an aluminum layer), and $(Li_{0.62}K_{0.38})CO_3$ molten electrolyte held within the membrane. Quantitatively, over 90% removal of H_2S has been demonstrated, although long-term transport performance at these levels is yet to be proven. Electrical polarization is also excellent, with far less than one volt needed at all currrents.

The removal rate of H_2S is limited by bulk mass transfer from the coal-gas stream to the cathode and migration of sulfur ions through the electrolytic membrane. Operating temperature and cell design parameters (i.e. membrane thickness, membrane tortuosity, and flow channel design) determine which

of these processes limits the maximum current allowable for H_2S removal. Higher temperatures favor higher migration rates while thicker, more tortuous membranes decrease migration rate.

Lost current efficiency due to side reactions are of little concern because the power density is small in this process; however, loss of electrolyte could be a concern if carbonate is extensively reacted to form CO_2 and O_2 at the anode. In this regard, testing is being carried out with clean coal-gas. These tests will assure that stoichiometric transport of CO_2 can be achieved in either direction; that is, in the standard fuel-cell mode, from cathode to anode, or in the concentration-cell mode, from anode to cathode. These results will also be shown.